High Resolution Electron Spectroscopy of Atomic Barium

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INTRODUCTION

High-resolution 4d and 5p photoelectron spectra of free barium atoms were measured using narrow bandwidth synchrotron radiation of 131.2 eV photon energy. These measurements were necessary to disentangle the complex structure of the electron spectra. We also performed a Hartree-Fock calculation with relativistic corrections. A very good agreement between theory and experiment was found, which allowed the assignment of all but the weakest lines in addition to pointing out the significant contribution of configuration interaction and shake processes.

EXPERIMENT

We The experiment was performed at the Atomic and Molecular beamline 10.0.1 of the Advanced Light Source synchrotron radiation facility at Lawrence Berkeley National Laboratory. The radiation from a 4.5 m long 10 cm period undulator was monochromatized by a spherical grating monochromator. A photon bandwidth of 40-50 meV was used with the 925 lines/mm grating at 131.2~eV photon energy. The electron spectra were measured using an end station designed for gas-phase angle-resolved studies, which is based on a Scienta SES-200 hemispherical analyzer \cite{Nora99}. The present measurements were all performed at the emission angle of 54.7º with respect to the electric field vector, in a plane perpendicular to the propagation direction of the linearly polarized photon beam. The analyzer was operated at the constant pass energy of 40 eV with an electron energy resolution of 40-50 meV. The kinetic energy scale of the spectrometer was calibrated by the Xe NOO Auger lines and the photon energy was determined using the binding energy of the Xe 4d photolines.

A resistively heated metal vapor oven was used to generate an effusive beam of Ba atoms. To increase the length of the interaction region, the nozzle of the stainless steel crucible contained eight channels (0.8 mm diam., 5 mm long) in a line, aligned parallel to the photon beam direction. A 1.5 mm thick μ-metal disk with a slit for the atomic beam was placed between the crucible and the interaction region to shield the magnetic field of the heater, crucible and other parts of the oven from the interaction region. On the other side of the interaction region, opposite to the crucible, a water cooled copper disk was mounted to collect and condense the metal vapor. The contact potential of the accumulating Ba on this disk caused a continuously changing field in the interaction region. To keep the highest possible electron energy resolution over a long period of time, the electron spectra were collected by one sweep at a time and added up later taking into account small shifts of a few meV. The operating temperature of the oven was in the range of 540-590ºC, at which the Ba vapor pressure inside the crucible was in the range of 1.5-5.0 mtorr.

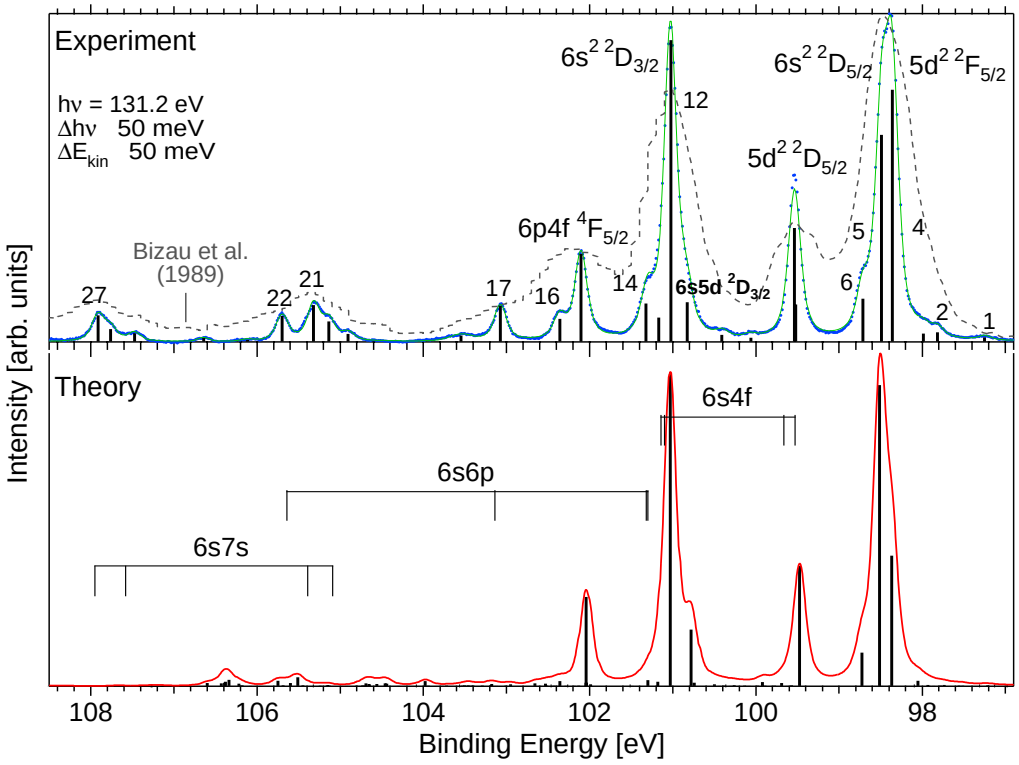

Figure 1. Barium 4d photoelectron spectra. (top) The solid line represents a fit with Voigt profiles. The dashed line is from Ref. [2]. (bottom) Calculated spectrum using the relativistic Hartree-Fock method. The vertical bars represent the fitted (top) and calculated (bottom) lines.

RESULTS

Our experimental and calculated spectra are shown in Fig. 1 together with previous data of Bizau et al. [2]. The measurements were performed with a photon bandwidth of 50 meV and a spectrometer resolution of 50 meV. In the strongest line (12) 66500 counts were collected during approximately 100 minutes of acquisition time. The spectrum was fitted with a set of Voigt profiles, where the lorentzian and gaussian widths (with the exception of lines 18,23,24) were kept the same for all lines. A natural linewidth of 113(10) meV and a gaussian broadening of 109(10) meV was obtained from the fit. The fitted positions and intensities of the peaks are indicated as vertical lines in the top panel of Fig. 1. The calculated lines are shown as vertical lines in the lower panel of Fig. 1. For better comparison the theoretical data has been convoluted with a 109 meV gaussian and a 113~meV lorentzian to match the experimental resolution and lifetime broadening, respectively.

The position and intensity of the calculated spectrum was matched to the measurement using line 12 by shifting the theoretical spectrum by 0.1 eV. For the main lines in the region of 98-102 eV binding energy, an excellent agreement is found for the line positions. The intensity distribution is also given rather well by the calculation, although some intensity is missing at 99.5 eV binding energy (lines 7,8) and the ratio of lines 4 and 5 is reversed. The satellite structure in this region is almost completely due to configuration interaction, which is taken into account in the present calculation. These lines can be assigned to the $4d^9\ 6s^2$, $4d^9\ 5d^2$, $4d^9$ 6s5d and $4d^9$ 6p4f states. Above 102 eV binding energy, considerable intensity is missing in the calculated spectrum. It is known from previous works [2,3], that the satellite lines above 102 eV are mostly due to the $4d^9$

6s7s shakeup and $4d^9$ 6s4f and $4d^9$ 6s6p conjugate shakeup processes. The missing intensity is expected for conjugate shake satellites because they cannot be taken into account in the present calculation. The normal shake satellites $4d^9$ 6s7s have been included via CI, but their estimated intensity is too low. To identify the experimentally observed lines, we determined the binding energy of the shake satellites from an electronic structure calculation. In the lower panel of Fig. 1 the positions of these satellites are marked as vertical bars. For simplification only the ^2D states are shown. The positions are in very good agreement with observed satellite lines. For the configuration 6s7s mixing with the 5d7s, which are located at lower binding energies, is most important. Neglecting this effect will result in about 0.3 eV too low binding energies. The influence of the comparable configurations 5d6p and 5d4f on the conjugate shake satellites 6s6p and 6s4f is much weaker. Nevertheless, the intensities of the satellites cannot be evaluated in this simple CI picture. This leads to the conclusion that the $4d^9$ 6s7s satellite lines are mainly populated by shake processes and not by final ionic state configuration interaction (FISCI), which is the main process for the $4d^9$ $5d^2$, $4d^9$ 6s5d and $4d^9$ 6p4f states. To estimate the intensities for the shake satellites we have calculated several overlap integrals as described above. A very good agreement is achieved for the normal shake-up intensities calculated in this approximation with the experimental data and also with the values given in Ref. [4]. However, this simple approximation fails for the conjugate shake process. This also explains the missing intensities in the theoretical spectrum at 99.5~eV and 101.2 eV binding energies. Thus lines 7 and 13 can be attributed to the 6s4f conjugate shakeup satellites.

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